

PATENT COOPERATION TREATY

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Basell Intellectual Property

- 2 Feb 2004

From the
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

BASELL POLYOLEFINE GMBH
 Intellectual Property
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PCT

NOTIFICATION OF TRANSMITTAL OF
THE INTERNATIONAL PRELIMINARY
EXAMINATION REPORT

(PCT Rule 71.1)

CB 2. Kms.

Date of mailing
(day/month/year)

30.01.2004

Applicant's or agent's file reference
LU6022/CB

IMPORTANT NOTIFICATION

International application No.
PCT/EP 03/05592International filing date (day/month/year)
28.05.2003Priority date (day/month/year)
12.06.2002Applicant
BASELL POLYOLEFINE GMBH et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

The applicant's attention is drawn to Article 33(5), which provides that the criteria of novelty, inventive step and industrial applicability described in Article 33(2) to (4) merely serve the purposes of international preliminary examination and that "any Contracting State may apply additional or different criteria for the purposes of deciding whether, in that State, the claimed inventions is patentable or not" (see also Article 27(5)). Such additional criteria may relate, for example, to exemptions from patentability, requirements for enabling disclosure, clarity and support for the claims.

Name and mailing address of the international
preliminary examining authority:

European Patent Office
 D-80298 Munich
 Tel. +49 89 2399 - 0 Tx: 523656 epmu d
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Authorized Officer

Brell, S

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PATENT COOPERATION TREATY

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INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference LU6022/CB	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/EP 03/05592	International filing date (day/month/year) 28.05.2003	Priority date (day/month/year) 12.06.2002
International Patent Classification (IPC) or both national classification and IPC C07F17/00		
Applicant BASELL POLYOLEFINE GMBH et al.		

1. This International preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 5 sheets, including this cover sheet.
 - This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 6 sheets.
3. This report contains indications relating to the following items:
 - I Basis of the opinion
 - II Priority
 - III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
 - IV Lack of unity of invention
 - V Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
 - VI Certain documents cited
 - VII Certain defects in the international application
 - VIII Certain observations on the international application

Date of submission of the demand 24.09.2003	Date of completion of this report 30.01.2004
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0.Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Richter, H Telephone No. +49 89 2399-8539



INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP 03/05592

I. Basis of the report

- With regard to the elements of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, Pages

1-40 as originally filed

Claims, Numbers

1-11 received on 12.01.2004 with letter of 09.01.2004

- With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- the language of publication of the international application (under Rule 48.3(b)).
- the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

- With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- contained in the international application in written form.
- filed together with the international application in computer readable form.
- furnished subsequently to this Authority in written form.
- furnished subsequently to this Authority in computer readable form.
- The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

- The amendments have resulted in the cancellation of:

- the description, pages:
- the claims, Nos.:
- the drawings, sheets:

- This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

- Additional observations, if necessary:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No.

PCT/EP 03/05592

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability;
citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes:	Claims	1-11
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-11
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-11
	No:	Claims	

2. Citations and explanations

see separate sheet

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP03/05592

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

Reference is made to the following documents:

D1 = RYABOV A N ET AL: 'ZIRCONIUM COMPLEXES WITH CYCLOPENTADIENYL LIGANDS INVOLVING FUSED A THIOPHENE FRAGMENT' ORGANOMETALLICS, ACS, COLUMBUS, OH, US, vol. 21, no. 14, 8 June 2002 (2002-06-08), pages 2842- 2855, XP001106373 ISSN: 0276-7333

D2 = Beilstein Registry Number 3261007

D3 = Beilstein Registry Number 3296411

D4 = EP-A-576970

D5 = EP-B- 790076

D1 describes cyclopentadienes (Cp'H) with fused thiophene and benzothiophene fragments:

4,5-dimethyl-6H- cyclopenta[b]thiophene (2), see scheme 1 5,6-dimethyl-4H-cyclopenta[b]thiophene (4), see scheme 2

2,3-dimethyl-1H-cyclopenta[b][1]benzothiophene (11), see scheme 5 1,2-dimethyl-3H-cyclopenta[b][1]benzothiophene (14). see scheme 14

Starting from the cyclopentadienes (2), (4), (11), and (14), several bis(cyclopentadienyl)dimethylsilanes were obtained and further used for the synthesis of the respective ansa-zirconocenes (see scheme 11), which were isolated and unambiguously characterized either as pure diastereomers or as rac/meso mixtures (28), (29), (31), and (32).

D1, column 1, last paragraph also mentions the use of the complexes in propene polymerization.

The ligands according to claim 4 as well as the transition metal complexes according to claim 1 are distinguished from the D1 ligands or complexes in that they are either bisindenyls or mixed indenyl/cyclopentathiophene or indenyl/ cyclopentapyrrol ligands or complexes. Hence independent claims 1 and 4 are novel over D1.

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT - SEPARATE SHEET**

International application No. PCT/EP03/05592

Claim 6 is novel over the Beilstein references D2 and D3 due to the definition of R3 and R4.

D4 which is now considered to be the closest prior art describes a bridged 4-phenyl indenyl metallocene (see claim 1). The positions 2 and 3 of the indenyl may be substituted by C1-C10-alkyl or C6-C10-aryl. As there is no example in D4 having that particular substitution pattern the overlap of D4 does not take away the novelty of claim 1 of the present application. This document only suggested the aforementioned substitution pattern. It is therefore (together with its divisional D5) the closest prior art in respect of the divalent group T and to the first divalent group T' as defined by claim 1. Subject-matter according to claims 1-11 is selected from D4; claims 1 and 6-11 and from D5 claims 6 (steps d-f), 8, and 9.

The asymmetric ligands and complexes according to claims 1, 3-5, and 7-11 are further away from D4 in that they are not available by selection from D4 alone. Rather they contain also structural elements disclosed in D1. As the 2,3-dialkyl-4-aryl-1-indenes according to D4 (see the description of the present application, page 1, lines 20-23) cannot directly be prepared claims 1-11, therefore, are inventive over D4 and D5 either alone or in combination with D1.

Art. 6 objections:

The second and the third options for T' according to claim 1 (thiophenyl and pyrrole derivatives) are not supported by way of examples. This subject-matter, therefore, does not meet the requirements of Art. 6, PCT and suffers under a-posteriori non unity due to D1. For easy understanding of the claims the definition of T should be combined with formula (I) in claims 1, 2 and 4

A reference to claim 1 should be added in claims 4 and 5 after formula (I).

07 decoy

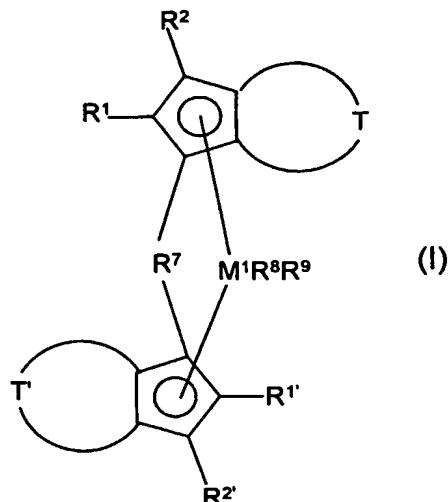
We claim:

1. A transition metal compound of the formula (I)

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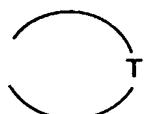
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where

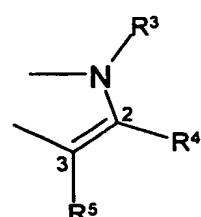
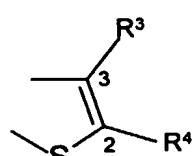
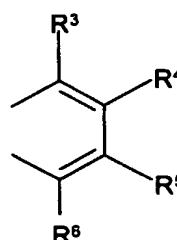
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is a divalent group such as

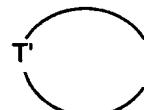
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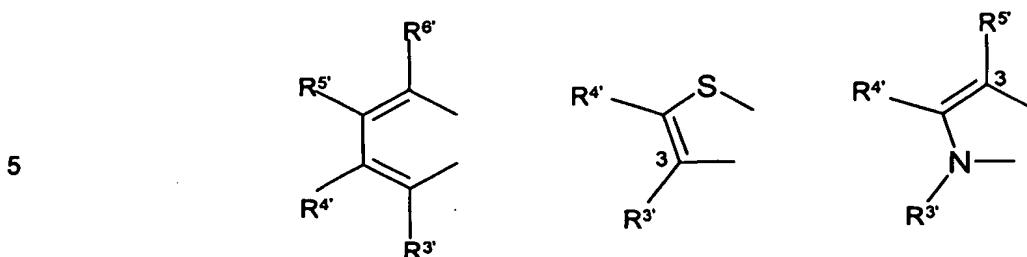
and

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is a divalent group such as

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and

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M^1 is titanium, zirconium or hafnium;

R^1, R^2 are identical or different and are each a C_1-C_{20} group;

R^1, R^2 are identical or different, identical to or different from R^1 or R^2 and are each hydrogen or a C_1-C_{20} group;

20

R^3 is a C_6-C_{18} -aryl group or C_4-C_{18} -heteroaryl; or a fluorinated C_6-C_{20} -aryl or C_7-C_{20} -alkylaryl, where the aryl part of these groups may bear one or more linear or branched C_1-C_{18} -alkyl, C_1-C_{18} -alkoxy, C_2-C_{10} -alkenyl or C_3-C_{15} -alkylalkenyl groups as substituents, or R^3 together with R^4 forms a monocyclic or polycyclic ring system which may in turn be substituted;

25

R^3' is hydrogen or a C_1-C_{40} group or R^3' together with R^4' forms a monocyclic or polycyclic ring system which may in turn be substituted;

R^4, R^4' are identical or different and are each hydrogen or a C_1-C_{20} group;

R^5, R^5', R^6, R^6' are identical or different and are each hydrogen or a C_1-C_{20} group;

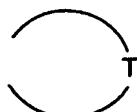
R^7 is a bridging structural element between the two indenyl radicals and is selected from the $M^2R^{10}R^{11}$ group, where M^2 is silicon, germanium, tin or carbon and R^{10} and R^{11} may be identical or different and are each hydrogen or a C_1-C_{20} -hydrocarbon-containing group;

30

R^8, R^9 may be identical or different and are each halogen, linear or branched C_1-C_{20} -alkyl, substituted or unsubstituted phenoxide, or R^8 and R^9 are joined to one another and form a monocyclic or polycyclic ring system which may in turn be substituted.

2. A transition metal compound as claimed in claim 1, wherein

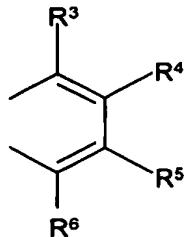
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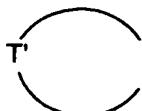
is

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and

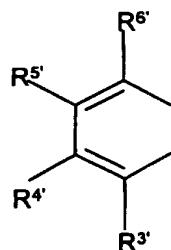
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is

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25

where the substituents R³ to R⁶ and R^{3'} to R^{6'} are defined as for formula (I).

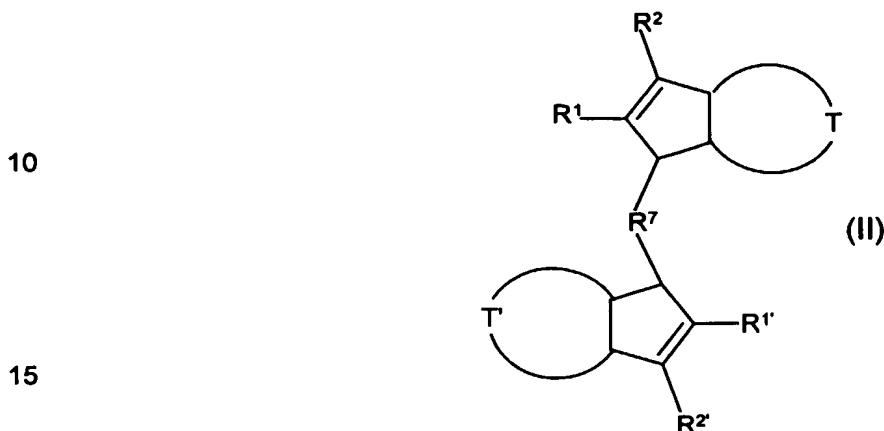
3. A transition metal compound as claimed in claim 1 or 2, wherein

M¹ is zirconium;R¹,R² are identical or different and are each a C₁-C₁₂-alkyl group;30 R^{1'},R^{2'} are identical or different and are each hydrogen, methyl, ethyl, n-propyl, isopropyl, n-butyl, isobutyl, tert-butyl, cyclopentyl or cyclohexyl;R³,R^{3'} are identical or different and are each a C₆-C₁₈-aryl group or two radicals R³ together with R⁴ and/or R^{3'} together with R^{4'} may form a monocyclic or polycyclic ring system which may in turn be substituted, and R^{3'} may also be hydrogen;35 R⁴,R^{4'} are identical or different and are either hydrogen or R⁴ together with R³ and/or R^{4'} together with R^{3'} form a monocyclic or polycyclic ring system;R⁵,R^{5'},R⁶,R^{6'} are identical or different and are each hydrogen, linear or branched C₁-C₁₈-alkyl, C₂-C₁₀-alkenyl or C₃-C₁₅-alkylalkenyl; C₆-C₂₀-aryl, C₄-C₁₈-heteroaryl, C₇-C₂₀-arylalkyl; or fluorinated C₁-C₁₂-alkyl, C₂-C₁₀-alkenyl, C₆-C₂₀-aryl or C₇-C₂₀-arylalkyl;

40

R^7 is a bridging structural element $SiR^{10}R^{11}$ and R^{10} and R^{11} are identical or different and are each a C₁-C₂₀-hydrocarbon-containing group and
 R^8, R^9 are each chlorine or methyl.

5 4. A ligand system of the formula (II) or its double bond isomers,



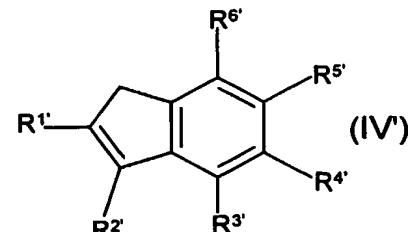
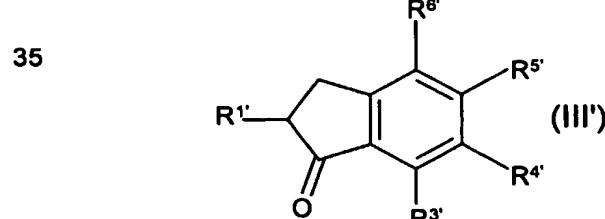
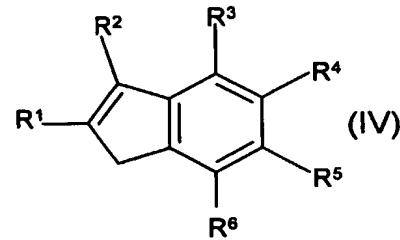
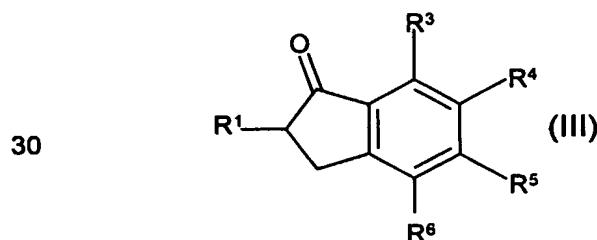
where the variables are as defined for formula (I).

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5. A process for preparing ansa-metallocenes of the formula (I), which comprises the following steps:

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a) reaction of a 1-indanone of the formula (III) or (III') with an organometallic compound $M^3R^{2m}Hal_n$ or $M^3R'^{2m}Hal_n$ and subsequent elimination to form the substituted indene of the formula (IV) or (IV'),

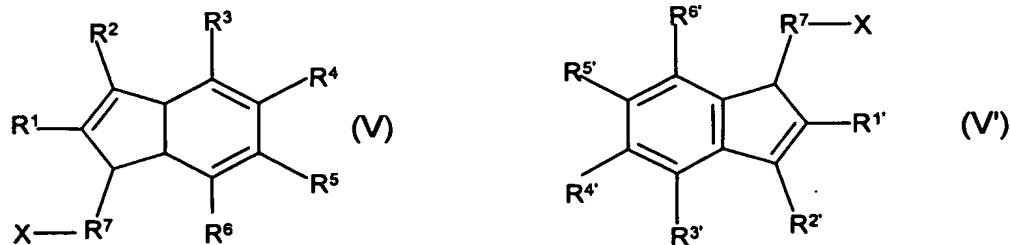


where the variables R¹, R^{1'}, R², R^{2'}, R³, R^{3'}, R⁴, R^{4'}, R⁵, R^{5'}, R⁶ and R^{6'} are as defined for formula (I), M³ is an alkali metal, an alkaline earth metal, aluminum or titanium, Hal is halogen, m is an integer and is equal to or greater than 1 and the sum of m+n corresponds to the valence of M³;

5

- b) deprotonation of the substituted indene of the formula (IV) or (IV') and subsequent reaction of the deprotonated indene with compounds of the type R⁷X₂ to form compounds of the formula (V) or (V') or their double bond isomers,

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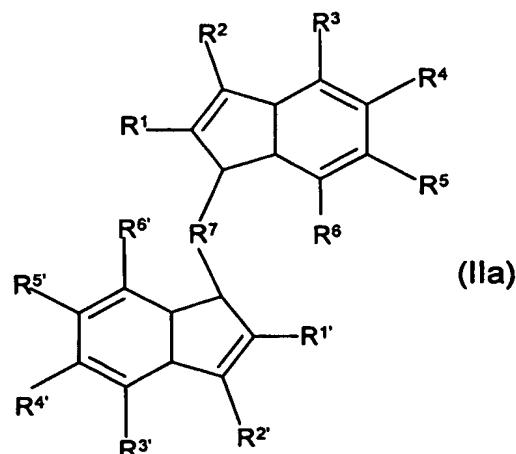
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where X is Cl, Br, I or O-tosyl and R⁷ is as defined for formula (I);

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- c) reaction of the compound of the formula (V) or (V') with a further deprotonated indene which has been obtained by deprotonation of (IV) or (IV') to form the ligand system of the formula (IIa) or its double bond isomers,

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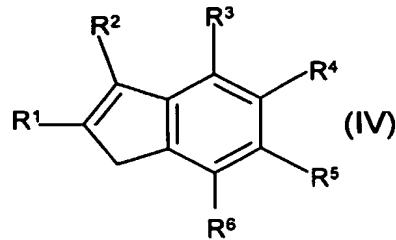
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- d) deprotonation of the ligand system of the formula (IIa) or its double bond isomers and reaction with compounds of the type X₂M¹R⁸R⁹ to give the ansa-metallocene of the formula (I), where X is as defined for formula (V) and M¹, R⁸ and R⁹ are as defined for formula (I).

6. An indene of the formula (IV) or its double bond isomer,

5



10 where the variables R^1 , R^2 , R^3 , R^4 , R^5 and R^6 are as defined for formula (I).

7. A catalyst system comprising one or more compounds of the formula (I) as claimed in any of claims 1 to 3 and one or more cocatalysts and/or supports.

15 8. The use of a catalyst system as claimed in claim 7 for the preparation of a polyolefin, in particular a copolymer of various olefins.

9. The use of a compound of the formula (I) as claimed in any of claims 1 to 3 for the preparation of a polyolefin, in particular a copolymer of various olefins.

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10. The use as claimed in claim 8 or 9 for the preparation of ethylene-propylene copolymers.

11. A process for preparing a polyolefin by polymerization of one or more olefins in the presence of one or more compounds of the formula (I) as claimed in any of claims 1 to 3.

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